

Cooperative resonance linewidth narrowing in a planar metamaterial

S. D. Jenkins and J. Ruostekoski

*School of Mathematics and Centre for Photonic Metamaterials,
University of Southampton, Southampton SO17 1BJ, United Kingdom*

(Dated: June 29, 2011)

We theoretically analyze the experimental observations of a spectral line collapse in a metamaterial array of asymmetric split ring resonators [Fedotov *et al.*, Phys. Rev. Lett. **104**, 223901 (2010)]. We show that the ensemble of closely-spaced resonators exhibits cooperative response, explaining the observed system-size dependent narrowing of the transmission resonance linewidth. We further show that this cooperative narrowing depends sensitively on the lattice spacing.

PACS numbers: 78.67.Pt,42.25.Bs,41.20.Jb,32.10.-f

Resonant multiple scattering plays an important role in mesoscopic wave phenomena that can also be reached with electromagnetic (EM) fields. In the strong scattering regime, interference of different scattering paths between discrete scatterers can result in, e.g., light localization [1]—an effect analogous to the Anderson localization of electrons in solids. Metamaterials comprise artificially structured media of plasmonic resonators interacting with EM fields. Due to several promising phenomena, such as the possibility for diffraction-free lenses resulting from negative refractive index [2], there has been a rapidly increasing interest in fabrication and theoretical modeling of such systems. The discrete nature of closely-spaced resonators in typical metamaterial arrays raises the possibility to observe also strong collective radiative effects in these systems.

In recent experiments Fedotov *et al.* observed a dramatic suppression of radiation losses in a 2D planar metamaterial array [3]. The transmission spectra through the metamolecular sheet was found to be strongly dependent upon the number of interacting metamolecules in the system. The transmission resonance quality factor increased as a function of the total number of active resonators, finally saturating at about 700 metamolecules. The metamaterial unit cell in the experiment was formed by an asymmetric split-ring (ASR) resonator, consisting of two circular arcs of slightly unequal lengths. The currents in these ASRs may be excited symmetrically (antisymmetrically), yielding a net oscillating electric (magnetic) dipole; Fig. 1.

In this letter we theoretically analyze the collective metamaterial response, observed experimentally in Ref. [3]. We find that strong interactions between a discrete set of resonators, mediated by the EM field, characterize the response of the ensemble and results in collective resonance linewidths and frequencies. We show how the cooperative response of sufficiently closely-spaced resonators is responsible for the observed dramatic narrowing of the transmission resonance linewidth (increasing quality factor) with the number of resonators [3]. In particular, the system exhibits a collective mode with an almost purely magnetic excitation, uniform phase profile,

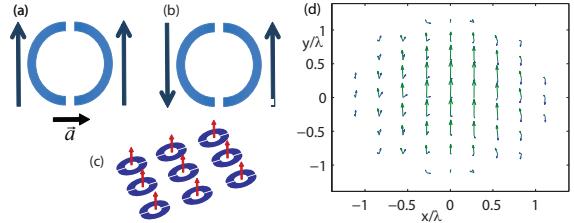


FIG. 1: (Color online) Asymmetric split ring metamolecules. (a) The symmetric mode with the currents in the two meta-atoms oscillating in-phase and (b) the anti-symmetric mode with the current oscillating π out-of-phase. For the symmetric case the dominant contribution is a net electric dipole moment in the plane of the resonator and for the anti-symmetric case a net magnetic dipole moment normal to the plane of the resonator. (c) The uniform phase-coherent collective magnetic eigenmode where all the metamolecules exhibit a magnetic dipole normal to the metamaterial plane. (d) Numerically calculated collective eigenmode v_m (for $u \simeq 0.28\lambda$, $\delta\omega \simeq 0.3\Gamma$). The amplitude and phase of magnetic (green) and electric (blue) dipoles are represented by the length and direction of the arrows, respectively.

and strongly suppressed radiative properties with each ASR possessing a nearly equal magnetic dipole moment. We show in detail how this mode can be excited by an incident plane wave propagating perpendicular to the array through an electric dipole coupling to an ASR, even when the magnetic dipole moments are oriented parallel to the propagation direction. We calculate the resonance linewidth of the phase-coherent collective magnetic mode that dramatically narrows as a function of the number of ASRs, providing an excellent agreement with experimental observations. The linewidth is also sensitive to the spacing of the unit cell resonators, with the closely-spaced ASRs exhibiting cooperative response, due to enhanced dipole-dipole interactions. Moreover, at the resonance, and with appropriately chosen parameters, nearly all the excitation can be driven into this mode. Due to its suppressed decay rate, the transmission spectrum displays a narrow resonance.

Our analysis demonstrates how essential features of the collective effects of the experiment in Ref. [3] can

be captured by a simple, computationally efficient model in which we treat each meta-atom as a discrete scatterer, exhibiting a single mode of current oscillation and possessing appropriate electric and magnetic dipole moments. Interactions with the EM field then determine the collective interactions within the ensemble. Moreover, our analysis indicates the necessity of accounting for the strong collective response of metamaterial systems and interference effects in multiple scattering between the resonators in understanding the dynamics and design of novel meta-materials. Strong interactions between resonators can find important applications in metamaterial systems, providing, e.g., precise control and manipulation of EM fields on a sub-wavelength scale [4], in developments of a lasing spaser [5], and disorder-related phenomena [6].

In order to analyze the experimental observations of the transmission spectra, we provide a simple theoretical model that is computationally efficient in large systems, which are required to generate strong collective effects, but that captures the essential features of interactions between the resonators. We consider an ensemble of N metamaterial unit elements, metamolecules, each formed by n discrete meta-atoms, with the position of the meta-atom j denoted by \mathbf{r}_j ($j = 1, \dots, n \times N$). This ensemble is driven by an external beam with electric field $\mathbf{E}_{\text{in}}(\mathbf{r}, t)$ and magnetic field $\mathbf{B}_{\text{in}}(\mathbf{r}, t)$ with wavelengths much larger than the size of the meta-atoms. Consequently, we treat meta-atoms as radiating dipoles and ignore the higher-order multipole-field interactions. We assume that each meta-atom j supports a single eigenmode of current oscillation with the charge $Q_j(t)$ as its dynamic variable. Then the associated electric and magnetic dipole moment for the meta-atoms are $\mathbf{d}_j = Q_j h_j \hat{\mathbf{d}}_j$ and $\mathbf{m}_j = I_j A_j \hat{\mathbf{m}}_j$, respectively, where $\hat{\mathbf{d}}_j$ and $\hat{\mathbf{m}}_j$ denote unit vectors and $I_j(t) = dQ_j/dt$ is the current. Here h_j and A_j are the corresponding proportionality coefficients (with the units of length and area), depending on the specific geometry of the resonators. In the dipole approximation the polarization and magnetization are given in terms of the density of electric and magnetic dipoles $\mathbf{P}(\mathbf{r}) = \sum_j \mathbf{P}_j(\mathbf{r})$ and $\mathbf{M}(\mathbf{r}) = \sum_j \mathbf{M}_j(\mathbf{r})$, where the polarization and the magnetization of the resonator j are $\mathbf{P}_j(\mathbf{r}, t) = \mathbf{d}_j \delta(\mathbf{r} - \mathbf{r}_j)$ and $\mathbf{M}_j(\mathbf{r}, t) = \mathbf{m}_j \delta(\mathbf{r} - \mathbf{r}_j)$, respectively. Here \mathbf{r}_j denotes the position of the meta-atom j .

Incident EM field drives the excitation of the current oscillations, generating an oscillating electric and magnetic dipole in each meta-atom. The resulting dipole radiation from the metamaterial array is the sum of the scattered electric and magnetic fields from all the meta-atoms $\mathbf{E}_S(\mathbf{r}, t) = \sum_j \mathbf{E}_{S,j}(\mathbf{r}, t)$ and $\mathbf{H}_S(\mathbf{r}, t) = \sum_j \mathbf{H}_{S,j}(\mathbf{r}, t)$, where $\mathbf{E}_{S,j}(\mathbf{r}, t)$ and $\mathbf{H}_{S,j}(\mathbf{r}, t)$ denote the electric and magnetic field emitted by the meta-atom j . The Fourier components of the scattered fields have the familiar expressions of electric and magnetic fields radiated by oscillating electric and magnetic dipoles [7].

We express these in terms of the positive frequency components $\mathbf{E}_S^+(\mathbf{r}, t)$ and $\mathbf{H}_S^+(\mathbf{r}, t)$ for the frequency Ω ($k \equiv \Omega/c$), where the total field $\mathbf{E}(\mathbf{r}) = \mathbf{E}^+(\mathbf{r}) + \mathbf{E}^-(\mathbf{r})$ and $(\mathbf{E}^+)^* = \mathbf{E}^-$,

$$\mathbf{E}_S^+(\mathbf{r}, \Omega) = \frac{k^3}{4\pi\epsilon_0} \int d^3r' \left[\mathbf{G}(\mathbf{r} - \mathbf{r}', k) \mathbf{P}^+(\mathbf{r}', \Omega) + \frac{1}{c} \mathbf{G}_\times(\mathbf{r} - \mathbf{r}', k) \mathbf{M}^+(\mathbf{r}', \Omega) \right], \quad (1)$$

$$\mathbf{H}_S^+(\mathbf{r}, \Omega) = \frac{k^3}{4\pi} \int d^3r' \left[\mathbf{G}(\mathbf{r} - \mathbf{r}', k) \mathbf{M}(\mathbf{r}', \Omega) - c \mathbf{G}_\times(\mathbf{r} - \mathbf{r}', k) \mathbf{P}^+(\mathbf{r}', \Omega) \right], \quad (2)$$

Here \mathbf{G} denotes the radiation kernel representing the electric (magnetic) field emitted from an electric (magnetic) dipole [7]. The explicit expression for the corresponding radiated field from a dipole with amplitude $\hat{\mathbf{v}}$ reads

$$\mathbf{G}(\mathbf{r}, k) \cdot \hat{\mathbf{v}} = (\hat{\mathbf{r}} \times \hat{\mathbf{v}}) \times \hat{\mathbf{r}} \frac{e^{ikr}}{kr} + [3\hat{\mathbf{r}}(\hat{\mathbf{r}} \cdot \hat{\mathbf{v}}) - \hat{\mathbf{v}}] \times \left[\frac{1}{(kr)^3} - \frac{i}{(kr)^2} \right] e^{ikr} - \frac{4\pi}{3} \delta(kr) \hat{\mathbf{v}}, \quad (3)$$

where $\hat{\mathbf{r}} \equiv \mathbf{r}/r$. Similarly, $\mathbf{G}_\times(\mathbf{r}, k)$ represents the radiation kernel for the magnetic (electric) field of an electric (magnetic) dipole source [7]. Specifically, the corresponding radiated field from a dipole with amplitude $\hat{\mathbf{v}}$ yields

$$\mathbf{G}_\times(\mathbf{r}, k) \cdot \hat{\mathbf{v}} = \frac{e^{ikr}}{kr} \left(1 - \frac{1}{ikr} \right) \hat{\mathbf{r}} \times \hat{\mathbf{v}}. \quad (4)$$

Each meta-atom interacts with the incident fields $\mathbf{E}_{\text{in}}(\mathbf{r}, t)$ and $\mathbf{H}_{\text{in}}(\mathbf{r}, t)$, as well as the fields radiated by all other meta-atoms in the metamaterial sample and with its own self-generated radiation. In each meta-atom the oscillating current generates electric and magnetic dipoles which both radiate magnetic and electric fields, according to Eqs. (1) and (2). These fields couple to dynamical variables of charge oscillations in other meta-atoms, producing more dipolar radiation. In order to calculate the EM response of the system, we solve the coupled set of equations involving all the resonators and the fields. A system of $n \times N$ single-mode resonators then possesses $n \times N$ collective modes of current oscillation. Each collective mode exhibits a distinct collective linewidth (decay rate) and resonance frequency, determined by the imaginary and real parts of the corresponding eigenvalue. The resulting dynamics resemble a cooperative response of atomic gases to resonant light in which case the EM coupling between different atoms is due to electric dipole radiation alone [8, 9]. The crucial component of the strong cooperative response of closely-spaced scatterers are *recurrent* scattering events [8, 10] – in which a wave is scattered more than once by the same dipole. Such processes cannot generally be modeled by the continuous medium electrodynamics, necessitating the meta-atoms to be treated as discrete scatterers. An approximate calculation of local field corrections

in a magnetodielectric medium of discrete scatterers was performed in Ref. [11] where the translational symmetry of an infinite lattice simplifies the response.

In this work we consider asymmetric metamolecules consisting of two meta-atoms with different resonance frequencies ω_j , centered around the frequency ω_0 , with $|\omega_j - \omega_0| \ll \omega_0$. The meta-atoms are illuminated by a near-resonant incident field with a narrow bandwidth and dominant frequency Ω_0 . For simplicity, we assume that the radiative electric and magnetic decay rates of each resonator j and sufficiently slow $\Gamma_{E/M} \ll \omega_0$. Then the dominant contribution of the dipole radiation in Eqs. (3) and (4) is generated at the wavenumber $k \simeq \Omega_0/c$ and a single isolated meta-atom behaves as a simple linear LC circuit with $\omega_j = 1/\sqrt{L_j C_j}$, where we have introduced an effective capacitance C_j and self-inductance L_j associated with the circuit current mode [12]. In the damping rate $\Gamma \equiv \Gamma_E + \Gamma_M + \Gamma_O$, we add to radiative dissipation also a nonradiative decay Γ_O , resulting, e.g., from ohmic losses. The interaction processes between the different resonators, mediated by dipole radiation and originating from Eqs. (1) and (2), are analogous to frequency dependent mutual inductance and capacitance, but due to the radiative long-range interactions, these can substantially differ from the quasi-static expressions for which \mathbf{G}_x is also absent.

In the transmission resonance experiment [3] an ASR formed a unit cell resonator (metamolecule). A single ASR consists of two separate coplanar circular arcs (meta-atoms), labeled by $j \in \{l, r\}$ and separated by $\mathbf{a} \equiv \mathbf{r}_r - \mathbf{r}_l$; Fig. 1. The current oscillations in each meta-atom produce electric dipoles with orientation $\hat{\mathbf{d}}$ ($\hat{\mathbf{d}} \perp \hat{\mathbf{a}}$) and magnetic dipoles with opposite orientations $\hat{\mathbf{m}}_r = -\hat{\mathbf{m}}_l$ ($\hat{\mathbf{m}}_r \perp \hat{\mathbf{a}}, \hat{\mathbf{d}}$). An asymmetry between the rings, in this case resulting from a difference in arc length, manifests itself as a difference in resonance frequencies with $\omega_r = \omega_0 + \delta\omega$ and $\omega_l = \omega_0 - \delta\omega$. In a single, isolated ASR the radiative interactions between the two resonators results in eigenstates analogous to superradiant and subradiant states in a pair of atoms. In order to analyze these eigenstates, we consider the dynamics of symmetric c_+ and antisymmetric c_- modes of current oscillation (Fig. 1) that represent the exact eigenmodes of the ASR in the absence of asymmetry $\delta\omega = 0$. Excitations of these modes possess respective net electric and magnetic dipoles and will thus be referred to electric and magnetic dipole excitations. The split ring asymmetry $\delta\omega \neq 0$, however, introduces an effective coupling between these modes in a single ASR, so that $\dot{c}_\pm = (-\gamma_\pm/2 \mp i\Delta)c_\pm - i\delta\omega c_\mp + F_\pm$, where γ_\pm and Δ denote the decay rates and a frequency shift, respectively, and F_\pm represents the driving by the incident field. Due to the asymmetry-dependent coupling between the modes, the incident field with $\mathbf{E}_{\text{in}} \parallel \hat{\mathbf{d}}$ and $\mathbf{B}_{\text{in}} \perp \hat{\mathbf{m}}$ only excites the electric mode when $\delta\omega = 0$, but for $\delta\omega \neq 0$ it

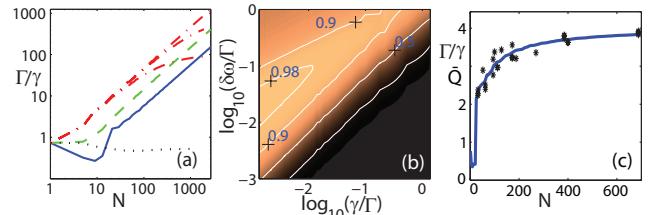


FIG. 2: (Color online) Cooperative metamaterial response displaying collective resonance narrowing. (a) The resonance linewidth γ of the collective magnetic mode v_m in the units of an isolated meta-atom linewidth Γ as a function of the number of metamolecules N ; the lattice spacings $u = 1/4\lambda$ (solid line), $3/8\lambda$ (dashed line), $1/2\lambda$ (dot-dashed line), and $3/2\lambda$ (dotted line). The intermediate dot-dashed line corresponds to an asymmetry $\delta\omega = 0.1\Gamma$, while $\delta\omega = 0$ for all other curves. The lower dot-dashed line incorporates nonradiative loss $\Gamma_O = 0.01\Gamma$ with all other curves assuming $\Gamma_O = 0$. (b) Overlap $O_m(b_f)$ of v_m with the state b_f excited by an incident field resonant on the mode v_m (for $u = 1/2\lambda$, and $\Gamma_O = 0$). For $\gamma \ll \Gamma$ there is a range of asymmetries $\delta\omega$ for which the incident field almost exclusively excites the mode v_m . (c) Comparison between experimentally measured transmission resonance quality factors $\bar{Q} = Q/Q_0$ (stars) from Ref. [3], where $Q_0 \simeq 4.5$ denotes the single ASR quality factor (for $\lambda \simeq 2.7\text{cm}$), and numerically calculated resonance linewidth γ of the collective magnetic mode v_m , both with $u \simeq 0.28\lambda$. Here $\delta\omega \simeq 0.3\Gamma$, $\Gamma_O \simeq 0.14\Gamma$.

can resonantly pump the anti-symmetric magnetic mode.

In order to model the experimentally observed collective response [3] we study an ensemble of identical ASRs (with $\mathbf{a} = a\hat{\mathbf{e}}_x$ and $\mathbf{d} = d\hat{\mathbf{e}}_y$; Fig. 1) arranged in a 2D square lattice within a circle of radius r_c , with lattice spacing u , and lattice vectors $(u\hat{\mathbf{e}}_x, u\hat{\mathbf{e}}_y)$. The sample is illuminated by a cw plane wave $\mathbf{E}_{\text{in}}^+(\mathbf{r}) = \frac{1}{2}\mathcal{E}\hat{\mathbf{e}}_y e^{i\mathbf{k}\cdot\mathbf{r}}$ with $\mathbf{k} = k\hat{\mathbf{e}}_z$, coupling to the electric dipole moments of the ASRs. In the experimentally measured transmission resonance through such a sheet [3] the number of active ASRs, N , was controlled by decoupling the ASRs with $r \gtrsim r_c$ from the rest of the system with approximately circular shaped metal masks with varying radii r_c . The resonance quality factor increased with the total number of active ASRs, saturating at about $N = 700$.

We find that an incident plane-wave drives all metamolecules uniformly and is phase-matched to collective modes in which the electric and/or magnetic dipoles oscillate in phase. In the absence of a split-ring asymmetry, only modes involving oscillating electric dipoles can be driven. These modes strongly emit perpendicular to the array (into the $\pm\hat{\mathbf{e}}_z$ directions) enhancing incident wave reflection. The magnetic dipoles, however, dominantly radiate into EM field modes within the ASR plane. This radiation may become trapped through recurrent scattering processes in the array, representing modes with suppressed emission rates and reflectance, and resulting in a transmission resonance. In order to quantify the effect, we study the radiation properties (for $\Gamma_E = \Gamma_M$) of

the numerically calculated collective magnetic eigenmode v_m of the system [Fig. 1(d)] which maximizes the overlap $O_m(b_A) \equiv |v_m^T b_A|^2 / \sum_i |v_i^T b_A|^2$ with the pure magnetic excitation $b_A = (1, -1, \dots, 1, -1)^T / \sqrt{2N}$ (all ASRs oscillating anti-symmetrically). We then show that the introduction of an asymmetry $\delta\omega$ in the resonances allows the excitation of v_m by the incident field. This mode closely resembles that responsible for the experimentally observed transmission resonance [3, 13].

Figure 2a shows dependence of the resonance linewidth γ of the collective mode v_m , which is the closest representation of the phase-coherent uniform magnetic excitation, on the number of metamolecules N for different lattice spacings u (λ denotes the resonance wavelength of v_m). In the absence of ohmic losses and for sufficiently small u and $\delta\omega$, $\gamma \propto 1/N$ for large N . The split ring asymmetry only weakly affects v_m . For $\delta\omega = 0.01\Gamma$, the curves representing γ are indistinguishable from those for $\delta\omega = 0$. For the relatively large $\delta\omega = 0.1\Gamma$, however, γ is increased for $N \gtrsim 200$. The cooperative response and linewidth narrowing sensitively depends on u . For larger u (e.g., $u = 3/2\lambda$), γ becomes insensitive to N , indicating the limit of independent scattering of isolated metamolecules and a diminished role of cooperative effects.

As with an isolated ASR, an asymmetry $\delta\omega \neq 0$ generates an effective two-step coupling between *collective* electric and magnetic modes: one leg of the transition is provided by the coupling of the incident field to collective electric dipole excitations, while the other is provided by the coupling introduced by the asymmetry. We illustrate this in Fig. 2(b), showing the relative population $O_m(b_f)$ of the resonantly driven collective magnetic mode v_m , where b_f is the state induced by the uniform driving resonant on v_m . We find that for $\gamma \ll \Gamma$ and $\delta\omega \gtrsim \gamma$, one can excite a state in which more than 98% of the energy is in the target mode v_m . For $\delta\omega \ll \gamma$, any excitation that ends up in v_m is radiated away before it can accumulate; the array behaves as a collection of radiating electric dipoles. For larger $\delta\omega$, the population of v_m decreases since the increased strength of the coupling between the symmetric and anti-symmetric modes begins to excite also other modes with nearby resonance frequencies. Although the density of modes which may be excited increases linearly with N , the corresponding reduction of γ means that a smaller $\delta\omega$ is needed to excite the target mode, and there is a range of asymmetries for which v_m is excited. The narrowing in γ combined with the near exclusive excitation of this mode implies that for larger arrays the radiation from the sheet is suppressed and hence the transmission enhanced as in Ref. [3]. In Fig. 2(c) we compare the experimentally observed transmission resonance [3] to our numerics for $\Gamma_O \simeq 0.14\Gamma$ that results in the expected saturation of quality factor with N . We use the experimental spacing $u \simeq 0.28\lambda$, when

$\delta\omega \simeq 0.3\Gamma$ is chosen consistently with the measurements in Ref. [13]. The excellent agreement of our simplified model that only includes dipole radiation contributions from each meta-atom can be understood by a notably weaker quadrupole than dipolar radiation field from an ASR [14]. The result also confirms the importance of the uniform magnetic mode v_m on the observed transmission resonance. The observed saturation is due to a combination of a fixed $\delta\omega$, which in larger arrays leads to the population of several other modes in addition to v_m , and ohmic losses in the resonators which set an ultimate limit to the narrowing of γ . Since in the experiment $\delta\omega \gtrsim \gamma$, the resonance narrowing could be improved by using a smaller $\delta\omega$, while a sufficient transfer of excitations to v_m can still be achieved; Fig. 2(b).

In conclusion, we analyzed the recent observations of transmission spectra in a metamaterial array of ASRs. We showed that the system can exhibit a strong cooperative response in the case of sufficiently closely-spaced resonators. Moreover, we demonstrated how an asymmetry in the split rings leads to excitation of collective magnetic modes by a field which does not couple directly to ASR magnetic moments. The excitation of this uniform phase-coherent mode results in cooperative response exhibiting a dramatic resonance linewidth narrowing, explaining the experimental findings [3].

We gratefully acknowledge N. Papasimakis, V. Fedotov, and N. Zheludev for discussions and providing the measurement data for the quality factor. This work was financially supported by the EPSRC.

- [1] D. S. Wiersma *et al.*, Nature **390**, 671 (1997).
- [2] D.R. Smith *et al.*, Science **305**, 788 (2004).
- [3] V. A. Fedotov *et al.*, Phys. Rev. Lett. **104**, 223901 (2010).
- [4] A. Sentenac and P. C. Chaumet, Phys. Rev. Lett. **101**, 013901 (2008); F. Lemoult *et al.*, *ibid.* **104**, 203901 (2010); T. S. Kao *et al.*, Phys. Rev. Lett. **106**, 085501 (2010).
- [5] N. I. Zheludev *et al.*, Nature Photonics **2**, 351 (2008).
- [6] N. Papasimakis *et al.*, Phys. Rev. B **80**, 041102(R) (2009).
- [7] J.D. Jackson, *Classical Electrodynamics*, (John Wiley & Sons, New York, 1998).
- [8] J. Ruostekoski and J. Javanainen, Phys. Rev. A **55**, 513 (1997); **56**, 2056 (1997); J. Javanainen *et al.*, Phys. Rev. A **59**, 649 (1999).
- [9] O. Morice *et al.*, Phys. Rev. A **51**, 3896 (1995).
- [10] A. Lagendijk and B. A. van Tiggelen, Phys. Rep. **270**, 143 (1996).
- [11] J. Kästel *et al.*, Phys. Rev. A **76**, 062509 (2007).
- [12] S. D. Jenkins and J. Ruostekoski, unpublished.
- [13] V. A. Fedotov *et al.* Phys. Rev. Lett. **99**, 147401 (2007).
- [14] N. Papasimakis, unpublished.